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(54) Title: DIELECTRIC HEATING OF THERMOPLASTIC COMPOSITIONS

(57) Abstract: A HF responsive composition is described comprising (A) a molecular sieve material, and (B) at least one interpolymer comprising (i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and (ii) polymer units derived from (a) at least one vinyl or vinylidene aromatic monomer, or (b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or (c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally (d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c). The HF responsive composition of the invention can be heated in a dielectric heating process and is useful, for example, to make articles of manufacture which are formed or assembled using HF welding technology.

DIELECTRIC HEATING OF THERMOPLASTIC COMPOSITIONS

FIELD OF THE INVENTION

This invention relates to polymer compositions which can be heated in a high frequency (HF), alternating electric field as well as structures and fabricated articles made from such compositions. More particularly, this invention concerns HF responsive thermoplastic polymer compositions comprising a molecular sieve material and at least one interpolymer comprising one or more aliphatic olefinic monomers and a vinyl or vinylidene aromatic monomer and/or a hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer.

BACKGROUND OF THE INVENTION

Dielectric heating provides a method of contactless, direct, volumetric heating of dielectric materials capable of absorbing electromagnetic energy and which are very poor electrical conductors. This is achieved by the application of an alternating electric field generated at a radio frequency (RF) of less than 300 megahertz (MHz), or microwave frequencies of above 300 MHz, to the dielectric material. The dielectric loss factor, also known as the dielectric loss index, is a measure of the energy loss in a material and indicative of how well a material can be heated in a high frequency, alternating electric field. Plasticized polyvinyl chloride (f-PVC) has a high dielectric loss factor (e.g., of 0.4 at 20 – 30 MHz for a flexible filled PVC composition) and responds well to dielectric heating. The dielectric loss factors of other widely used thermoplastic polymers, including polyethylene, polystyrene or polypropylene, are known to be far too low to permit efficient dielectric heating (e.g. 0.0008 for polyethylene and 0.001 for polystyrene and polypropylene, respectively; see e.g. T.L. Wilson, at page 11 of "Dielectric Heating" in The Encyclopedia of Polymer Science and Engineering, Vol. 5, 2nd Ed., John Wiley & Sons, New York, 1989).

The dielectric heating process is clean, controllable and eliminates problems associated with the application of direct heat. The process also is economically attractive for use in commercial or industrial manufacturing processes provided that the employed materials are suitably lossy. Dielectric heating can advantageously be used in polymer processing. RF welding or RF sealing, also referred to as high frequency (HF) welding or

HF sealing, is a well established method for integrally joining suitable plastic materials under controlled conditions. Up to today, PVC or polyvinylidene chloride (PVDC), are preferred polymers in the manufacture of thermoplastic products involving a dielectric heating step, in particular a HF welding step. However, due to the growing concern about the environmental impact and other drawbacks associated with the use of these polymers, the interest for suitable alternative materials has significantly increased. One desideratum for any potential substitute material is HF responsiveness, in particular HF weldability, since the existing processing lines typically include HF welding facilities. Over the past years ethylene- and propylene-based polymer compositions have gained considerable attention as alternatives for PVC or PVCD polymers. While structures and articles made from polyethylene or polypropylene can be thermally welded or heat-sealed, these polymers do not exhibit adequate dielectric properties to permit efficient dielectric heating in general or HF welding in particular.

The prior art reports various attempts for making HF responsive polyolefin compositions. For example, one approach uses polar copolymers of olefins, e.g. copolymers of ethylene and an acrylate ester or vinyl acetate (VA). Copolymers of ethylene and higher levels of vinyl acetate or methyl acrylate have been found to exhibit some RF activity (see e.g. UK Patent Application No. 2 177 974). Blends comprising ethylene/styrene interpolymers as major component and ethylene vinyl acetate (EVA) or thermoplastic polyurethane (TPU) as minor component are mentioned to have HF weldability (C.F. Diehl et al., 57th SPE ANTEC (1999), pages 2149 – 2153). International Patent Application WO 94/12569 reports good RF sealing properties for films made from blends comprising a propylene-ethylene copolymer and an ethylene-alkyl acrylate copolymer. International Patent Application WO 00/69629 provides RF weldable films made from a blend of a non-polar olefin polymer and a polar copolymer with carbon monoxide (CO).

According to U.S. Patent No. 3,336,173, RF weldable polyethylene and polypropylene compositions are obtained by the incorporation of long chain synthetic polyamide resin. U.S. Patent No. 6,136,923 provides thermoplastic compositions of alpha-olefin monomers with one or more vinylidene aromatic monomers and/or one or more hindered aliphatic or cycloaliphatic vinylidene monomers blended with polyvinyl chloride. It is mentioned that such compositions can be RF sealable.

Japanese Patent Application No. 10-219048 describes RF weldable polypropylene compositions consisting of fine metal oxide particles, ion-substituted zeolite and polypropylene. The ion-substituted zeolite, in which a portion or all of the exchangable ions are replaced with ammonium ions and silver ions, is used as nucleating agent and chlorine scavenger. European Patent Application No. 193 902 proposes to use zinc oxide, bentonite clay, or crystalline and amorphous alkali or alkaline earth metal alumino silicates as radio frequency sensitizers for high molecular weight high density polyethylene or ultrahigh molecular weight polyethylene. It is taught that the radio frequency sensitizers have to be essentially dried before use. European Patent No. 149 782 discloses compositions comprising silane-grafted polyolefins and a crystalline zeolite. The compositions are shapeable into articles which can be cross-linked after shaping by exposure to humidity and/or microwaves.

There still is the need for polymer compositions which are suitable as substitute materials for PVC, especially flexible PVC, or PVCD and which can be dielectrically heated efficiently. In particular, there is a need for polymer compositions which are HF responsive without depending on the presence of HF active amounts of other polymers, such as, for example, ethylene/vinyl acetate (EVA) copolymer, ethylene-alkyl acrylate copolymer, thermoplastic polyurethane (TPU) or PVC to impart HF responsiveness. Furthermore, there is a need for such polymer compositions which can be formed into RF weldable structures, including, for example, a film, a foam, a profile, fibers or a molding, or a fabricated article.

It is one object of the present invention to meet these and other needs. In particular it is an object of the present invention to provide new HF responsive compositions with favorable properties. Such compositions provide, overall, an advantageous balance of properties as compared to those materials which have been heretofore known in the art or commercially used. It is a particular object of the present invention to provide polymer compositions which are HF weldable and have a good balance of mechanical properties. There are a wide range of polymer structures and fabricated articles which could benefit from being prepared from new and improved HF responsive thermoplastic polymer compositions. Such polymer structures include, but are not limited to films, foams, fibers, profiles and molded materials and fabricated articles which are assembled by the HF methods.

Very specifically, it is an object of the present invention to provide compositions and structures comprising at least one interpolymer comprising at least one aliphatic olefinic monomer and at least one vinyl or vinylidene aromatic monomer and/or at least one hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer which can be processed on conventional HF welding equipment under commercially acceptable conditions.

SUMMARY OF THE INVENTION

The present invention relates to HF responsive compositions comprising

(A) a molecular sieve material, and

(B) an interpolymer comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c).

In another aspect, the invention relates to a structure comprising a HF responsive composition provided herein which has undergone a film-forming, a fiber-forming, a foam-forming, a profile-forming, a coating or a molding process.

Another aspect of the present invention is a fabricated article comprising or made from a HF responsive composition or structure provided herein.

DETAILED DESCRIPTION OF THE INVENTION

Definitions

All references herein to elements or metals belonging to a certain Group refer to the Periodic Table of the Elements published and copyrighted by CRC Press, Inc., 1989. Also,

any reference to the Group or Groups shall be to the Group or Groups reflected in this Periodic Table of the Elements using the IUPAC system for numbering groups.

The term "HF responsive" in the context of a composition of the present invention means that such composition can be heated by exposure to a time varying electric field generated at a frequency in the range of from 0.1 – 30,000 MHz.

The term " molecular sieve" as used herein means an inorganic solid and porous material which is capable of acting as a sieve on a molecular basis.

The term "structure" as used herein in the context of "structure made from a HF responsive composition according to the present invention" is defined as a composition of the present invention which has undergone a one step film-forming, fiber-forming, foam-forming, profile-forming or molding process.

The term "copolymer" as employed herein means a polymer wherein two different monomers are polymerized to form the copolymer.

The term "interpolymer" is used herein to indicate a polymer wherein at least two different monomers are polymerized to make the interpolymer. This includes, for example, copolymers, terpolymers and the like.

The term "derived from" means made or mixed from the specified materials, but not necessarily composed of a simple mixture of those materials. Compositions "derived from" specified materials may be simple mixtures of the original materials, and may also include the reaction products of those materials, or may even be wholly composed of reaction or decomposition products of the original materials.

The term "comprising" as used herein means "including".

All parts and percentages are by weight unless indicated otherwise.

Unless otherwise stated, any given range includes both endpoints used to state the range.

Surprisingly, it has been found that a molecular sieve material provides for HF activity or HF responsiveness of compositions comprising at least one interpolymer comprising one or more aliphatic olefinic monomers and one or more vinyl or vinylidene aromatic monomers and/or one or more hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers. Such HF-responsive compositions can be heated via a dielectric heating process resulting in localized softening or melting of the interpolymers. The

compositions of the invention have sufficient dielectric loss properties to convert the HF energy to thermal energy. Preferably, the HF responsiveness of the compositions provided by the present invention is such that structures comprising or made from said compositions can be efficiently HF welded by the application of dielectric heating at the location of weld.

Component (A) characterizing the compositions of the present invention is composed of one or more molecular sieve materials. Such molecular sieve materials are inorganic solid materials characterized by a porous structure which is formed by channels or pores. The pores may be nonuniform with varying pore diameters, or uniform with constant pore diameters. The porous structure provides for a large internal surface area with adsorptive capacity for molecular species which are capable of entering the pores, e.g. water molecules. The pores may penetrate essentially the entire volume of the solid material. Molecular sieve materials may be amorphous or crystalline. Examples of molecular sieve materials suitable for the purpose of the present invention include, without limitation, clays, silica, zeolites, aluminophosphates, metalloaluminophosphates, silicoaluminophosphates, metallosilicophosphates and sulfide-based framework structures. Unless expressly stated otherwise, the terms “molecular sieve”, “molecular sieve material” or any such specific material, e.g. zeolite, are used herein independently from the adsorption state, e.g. the hydration state, of the porous material. For example, the molecular sieve material may be fully hydrated, partially hydrated or dehydrated.

Suitable clays include, for example, montmorellite clay, and are readily commercially available, e.g. from Suedchemie, Germany.

Suitable silica materials include, for example, synthetic amorphous precipitated silicas (Chemical Abstracts Registration Number 112926-00-8), which are readily available commercially, e.g. the Sipernat® silicas available from Degussa Huels, Germany.

Preferred are molecular sieve materials which have an ordered crystalline structure providing pores of a substantially uniform diameter. The pores may have diameters in the micro-size range of below about 2 nm or in the meso-size range of from about 2 nm to about 20 nm. Preferably, the pores are contained within the crystalline structure rather than formed by the spaces between randomly agglomerated crystals. For the purpose of the present invention, zeolites are the most preferred molecular sieve materials.

Zeolites are synthetic or natural crystalline silicates or aluminosilicates with channels of diameters of less than 1.2 nm. Zeolites may also be referred to as framework aluminosilicates, because the framework is based on a three-dimensional network of $[(Al,Si)O_4]$ tetrahedra which are linked to each other (at the corners) by shared oxygen atoms. Substitution of aluminum for silicon generates a charge imbalance, requiring the inclusion of a cation. In the hydrated form, zeolites of the most important aluminosilicate variety may be represented by the formula:

$$M_{x/n}[(AlO_2)_x(SiO_2)_y] \bullet wH_2O,$$

wherein M represents a cation of valence n, x is the number of aluminum-based tetrahedra and charge balancing cations in the unit cell, y is the number of silicon-based tredrahedra in the unit cell, and w is the number of water molecules in the zeolite pores. The bracketed part of the formula $[(AlO_2)_x(SiO_2)_y]$ represents the anionic framework composition. The sum $(x + y)$ is the total number of tetrahedra in the unit cell. The ratio y/x may vary depending on the type of zeolite and typically has values of from about 1 to about 6 depending upon the structure. Occlusion of AlO_2^- anions may result in a ratio of below 1. The framework contains channels and voids which can be occupied by the cations M and water molecules. The water molecules may be reversibly removed, for example, by the application of heat. The cation M may be a Group IA element or a Group IIA element, that is it may be an alkali metal cation or an alkaline-earth metal cation. Typically, M is selected from the group consisting of sodium, potassium, magnesium, calcium, strontium and barium. In synthetic zeolites, ammonium and/or alkylammonium cations, e.g. $CH_3NH_3^+$, $(CH_3)_2NH_2^+$, $(CH_3)_3NH^+$ or $(CH_3)_4N^+$ cations, may be incorporated. In some synthetic zeolites, aluminum cations are substituted by gallium cations, or silicon cations may be substituted by germanium or phosphorus cations, incorporation of the latter necessitating a modification of the structural formula. Detailed teachings regarding zeolite structures and properties are seen in Donald W. Breck, "Zeolite Molecular Sieves – Structure, Chemistry, And Use", copyright 1974, by John Wiley & Sons, Inc.

Natural zeolite minerals suitable for use in the present invention include, for example, chabazite, mordenite, and ferrierite. Suitable synthetic zeolites include those within the families of Type A, Type X, Type Y, Type L, Type omega and Type ZSM-5. Such zeolites are readily commercially available from numerous suppliers, for example from UOP, Tosoh, Grace Davison, Fluka Chemika, Switzerland (e.g. UOP 4A® zeolite type

4A), CU Chemie, Uetikon, Switzerland (e.g. PURMOL® 3 ST zeolite type 3A, PURMOL® 4ST zeolite type 4A, PURMOL® 5 zeolite type 5A and PURMOL® 13 zeolite type 13X), Tricat Zeolites, Bitterfeld, Germany (e.g. ZeoMax® 2210P zeolite type 4A), and Crosfield, Eijsden, Netherlands (e.g. Doucil® 4A zeolite type 4A). The specifically exemplified zeolite grades have a pore size in the range of from about 0.3 nm to about 0.8 nm and a particle size in the range of from about 2 microns (μm) to about 16 μm. For the purpose of the present invention, the zeolites may advantageously be used as (commercially) supplied, in particular in the supplied form and hydration state. Prior treatment or activation, e.g. to remove adsorbed water, is not considered necessary. Advantageously, the zeolites are used in powder form.

It is within the scope of the present invention that component (A) is composed of two or more molecular sieve materials, e.g. a mixture of two or more different molecular sieve materials, such as a mixture of different types of zeolites.

The interpolymer or interpolymers composing component (B) of the compositions of the present invention are thermoplastic interpolymers comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c).

Suitable aliphatic olefin monomers under (i) include ethylene and alpha-olefins (α-olefins), for example, α-olefins containing from 3 to about 20 carbon atoms, preferably from 3 to 12, more preferably from 3 to 8 carbon atoms. These α-olefins do not contain an aromatic moiety. Particularly suitable aliphatic olefin monomers are ethylene, propylene,

butene-1, 4-methyl-1-pentene, hexene-1 or octene-1, or ethylene in combination with one or more of propylene, butene-1, 4-methyl-1-pentene, hexene-1, or octene-1.

Suitable vinyl or vinylidene aromatic monomers within the meaning of (ii) (a) or (ii) (c), above, include, for example, those represented by the following formula:

$$\begin{array}{c} \mathrm{Ar} \\ | \\ (\mathrm{CH_2})_n \\ | \\ R^1 - \mathrm{C} = \mathrm{C}(R^2)_2 \end{array}$$

wherein R^1 is selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; each R^2 is independently selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; Ar is a phenyl group or a phenyl group substituted with from 1 to 5 substituents selected from the group consisting of halo, C_1-C_4-alkyl, and C_1-C_4-haloalkyl; and n has a value from zero to about 4, preferably from zero to 2, most preferably zero. Exemplary vinyl aromatic monomers include styrene, vinyl toluene, α-methylstyrene, t-butyl styrene, chlorostyrene, including all isomers of these compounds, and the like. Particularly suitable vinyl aromatic monomers include styrene and lower alkyl- or halogen-substituted derivatives thereof. Preferred monomers include styrene, α-methyl styrene, the lower alkyl-(C_1-C_4) or phenyl-ring substituted derivatives of styrene, such as for example, ortho-, meta-, and para-methylstyrene, the ring halogenated styrenes, para-vinyl toluene or mixtures thereof, and the like. The most preferred aromatic vinyl monomer is styrene.

Suitable hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds within the meaning of (ii) (b) or (c) are addition polymerizable vinyl or vinylidene monomers corresponding to the formula:

$$\begin{array}{c} A^1 \\ | \\ R^1 - \mathrm{C} = \mathrm{C}(R^2)_2 \end{array}$$

wherein A^1 is a hindered, aliphatic or cycloaliphatic substituent of up to 20 carbons, R^1 is selected from the group of radicals consisting of hydrogen and alkyl radicals containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; each R^2 is independently selected from the group of radicals consisting of hydrogen and alkyl radicals

containing from 1 to 4 carbon atoms, preferably hydrogen or methyl; or alternatively R^1 and A^1 together form a ring system.

The term "hindered" denotes that the monomer bearing this substituent is normally incapable of addition polymerization by standard Ziegler-Natta polymerization catalysts at a rate comparable with ethylene polymerizations. The term is used in the sense of "sterically bulky" or "sterically hindered". Aliphatic α-olefins having a simple linear structure including, for example, propylene, butene-1, 4-methyl-1-pentene, hexene-1 or octene-1 are not considered as hindered aliphatic monomers.

Preferred hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds are monomers in which one of the carbon atoms bearing ethylenic unsaturation is tertiarily or quaternarily substituted. Examples of such substituents include cyclic aliphatic groups such as cyclohexyl, cyclohexenyl, cyclooctenyl, or ring alkyl or aryl substituted derivatives thereof, tert-butyl, norbornyl, and the like. Most preferred hindered aliphatic or cycloaliphatic vinyl or vinylidene compounds are the various isomeric vinyl-ring substituted derivatives of cyclohexene and substituted cyclohexenes, and 5-ethylidene-2-norbornene. Especially suitable are 1-, 3-, and 4-vinylcyclohexene.

If the substantially random interpolymer contains a vinyl or vinylidene aromatic monomer and a sterically hindered aliphatic or cycloaliphatic monomer in polymerized form according to (ii) (c), the weight ratio between these two monomer types is not critical. Preferably, the interpolymer component (B) comprises polymer units (ii) derived from either (a) one or more vinyl or vinylidene aromatic monomers, or (b) one or more hindered aliphatic or cycloaliphatic monomers. Vinyl or vinylidene aromatic monomers (a) are preferred over hindered aliphatic or cycloaliphatic monomers.

Optional other polymerizable ethylenically unsaturated monomers (d) include strained ring olefins such as norbornene and C_1-C_{10} alkyl or C_6-C_{10} aryl substituted norbornenes. Further, one or more dienes can optionally be incorporated into the interpolymer to provide functional sites of unsaturation on the interpolymer useful, for example, to participate in crosslinking reactions. While conjugated dienes such as butadiene, 1,3-pentadiene (that is, piperylene), or isoprene may be used for this purpose, nonconjugated dienes are preferred. Typical nonconjugated dienes include, for example the open-chain nonconjugated diolefins such as 1,4-hexadiene (see U.S. Patent No. 2,933,480) and 7-methyl-1,6-octadiene (also known as MOCD); cyclic dienes; bridged ring cyclic

dienes, such as dicyclopentadiene (see U.S. Patent No. 3,211,709); or alkylidene-norbornenes, such as methylenenorbornene or ethylidenenorbornene (see U.S. Patent No. 3,151,173). The nonconjugated dienes are not limited to those having only two double bonds, but rather also include those having three or more double bonds. The diene may be incorporated in the interpolymer component (B) in an amount of from 0 to 15 weight percent based on the total weight of the interpolymer.

The interpolymer component (B) may consist of one, two, three, four or more interpolymers, e.g. component (B) may be formed by a blend of interpolymers.

Preferred interpolymers for use as component (B) in the present invention are interpolymers comprising one or more vinyl aromatic monomers in combination with ethylene, or a combination of ethylene and one or more $C_3 - C_8$ alpha olefin monomers, or a combination of ethylene and norbornene. Particularly preferred interpolymers include those selected from the group consisting of ethylene/styrene, ethylene/propylene/styrene, ethylene/butene/styrene, ethylene/pentene/styrene, ethylene/hexene/styrene, ethylene/octene/styrene or ethylene/styrene/norbornene.

The interpolymers comprising polymer units defined above under (a), (b), (c) and optionally (d) can be substantially random, pseudo-random, random, alternating, diadic, triadic, tetradic or any combination thereof. That is, the interpolymer product can be variably incorporated and optionally variably sequenced. The preferred sequence is substantially random. The preferred substantially random interpolymers are the so-called pseudo-random interpolymers as described in EP-A-0 416 815 by James C. Stevens et al. and US Patent No. 5,703,187 by Francis J. Timmers, both of which are incorporated herein by reference in their entirety. The preferred substantially random interpolymers also include the interpolymers of ethylene, olefinic monomers and vinyl aromatic monomers as described in US Patent No. 5,872,201 by Yunwa W. Cheung et al., which is incorporated herein by reference in its entirety.

The term "variably incorporated" as used herein refers to an interpolymer, particularly an ethylene/styrene interpolymer, manufactured using at least two catalyst systems wherein during interpolymerization the catalyst systems are operated at different incorporation or reactivity rates. For example, the interpolymer product having a total styrene content of 36 weight percent is variably incorporated where one catalyst system incorporates 22 weight percent styrene and the other catalyst system incorporates 48 weight

percent styrene and the production split between the two catalyst systems is 47/53 weight percentages.

"Random" interpolymers are those in which the monomer units are incorporated into the chain such that there exist various combinations of ordering including blockiness, e.g. where either the ethylene or the aliphatic alpha-olefin monomer or the sterically hindered vinyl or vinylidene monomer, or both can be repeated adjacent to one another.

Representative "alternating" interpolymers are, for example, alternating ethylene/styrene interpolymers in which the ethylene and the sterically hindered vinylidene monomer occur in repeat alternate sequences on the polymer chain in atactic or stereospecific structures (such as isotactic or syndiotactic) or in combinations of the general formula $(AB)_n$.

The term "substantially random" as used herein in reference to the interpolymers comprising the above-mentioned monomers (a), (b), (c) and optionally (d), and to ethylene/ styrene interpolymers in particular, generally means that the distribution of the monomers composing the interpolymer can be described by the Bernoulli statistical model or by a first or second order Markovian statistical model, as described by J. C. Randall in Polymer Sequence Determination, Carbon-13 NMR Method, Academic Press New York, 1977, pp. 71-78, the disclosure of which is incorporated herein by reference. Substantially random interpolymers do not contain more than 15 mole percent of the total amount of vinyl or vinylidene aromatic monomer in blocks of vinyl or vinylidene aromatic monomer of more than 3 units.

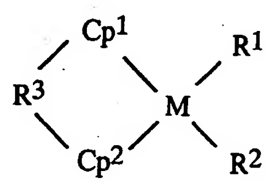
Preferably, the substantially random interpolymer is not characterized by a high degree (greater than 50 mole percent) of either isotacticity or syndiotacticity. This means that in the carbon-13 NMR spectrum of the substantially random interpolymer, the peak areas corresponding to the main chain methylene and methine carbons representing either meso diad sequences or racemic diad sequences should not exceed 75 percent of the total peak area of the main chain methylene and methine carbons.

A preferred method of preparation of the substantially random interpolymers includes polymerizing a mixture of polymerizable monomers in the presence of one or more metallocene or constrained geometry catalysts in combination with various cocatalysts, as described in EP-A-0,416,815 by James C. Stevens et al. and US Patent No. 5,703,187 by Francis J. Timmers, both of which are incorporated herein by reference in their entirety.

Preferred operating conditions for such polymerization reactions are pressures from atmospheric up to 3000 atmospheres and temperatures from -30°C to 200°C. Polymerizations and unreacted monomer removal at temperatures above the autopolymerization temperature of the respective monomers may result in formation of some amounts of homopolymer polymerization products resulting from free radical polymerization.

Examples of suitable catalysts and methods for preparing the substantially random interpolymers are disclosed in EP-A-514,828; as well as U.S. Patents: 5,055,438; 5,057,475; 5,096,867; 5,064,802; 5,132,380; 5,189,192; 5,321,106; 5,347,024; 5,350,723; 5,374,696; 5,399,635; 5,470,993; 5,703,187; and 5,721,185, all of which patents and applications are incorporated herein by reference.

The substantially random $C_2 - C_{20}$ olefin/vinyl aromatic monomer interpolymers can also be prepared by the methods described in JP 07/278230 employing compounds shown by the general formula



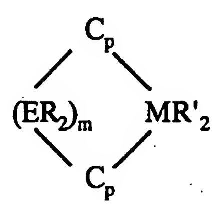
wherein Cp^1 and Cp^2 are cyclopentadienyl groups, indenyl groups, fluorenyl groups, or substituents of these, independently of each other; R^1 and R^2 are hydrogen atoms, halogen atoms, hydrocarbon groups with carbon numbers of 1-12, alkoxyl groups, or aryloxyl groups, independently of each other; m is a group IV metal, preferably Zr or Hf, most preferably Zr; and R^3 is an alkylene group or silanediyl group used to cross-link Cp^1 and Cp^2.

The substantially random aliphatic olefin/vinyl aromatic (co-)monomer interpolymers can also be prepared by the methods described by John G. Bradfute et al. (W. R. Grace & Co.) in WO 95/32095; by R. B. Pannell (Exxon Chemical Patents, Inc.) in WO 94/00500; and in Plastics Technology, p. 25 (September 1992), all of which are incorporated herein by reference in their entirety.

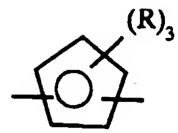
Also suitable are the substantially random interpolymers which comprise at least one aliphatic olefin/vinyl aromatic/vinyl aromatic/aliphatic olefin tetrad disclosed in WO

98/09999 by Francis J. Timmers et al.. These interpolymers contain additional signals in their carbon-13 NMR spectra with intensities greater than three times the peak to peak noise. These signals appear in the chemical shift ranges of 43.70 - 44.25 ppm and 38.0 - 38.5 ppm. Specifically, major peaks are observed at 44.1, 43.9, and 38.2 ppm. A proton test NMR experiment indicates that the signals in the chemical shift region 43.70 - 44.25 ppm are methine carbons and the signals in the region 38.0 - 38.5 ppm are methylene carbons. It is believed that these new signals are due to sequences involving two head-to-tail vinyl aromatic monomer insertions preceded and followed by at least one aliphatic olefin insertion, e.g. an ethylene/styrene/styrene/ethylene tetrad wherein the styrene monomer insertions of said tetrads occur exclusively in a 1,2 (head to tail) manner. It is understood by one skilled in the art that for such tetrads involving a vinyl aromatic monomer other than styrene and an α-olefin that the α-olefin /vinyl aromatic monomer/vinyl aromatic monomer/α-olefin tetrad will give rise to similar carbon-13 NMR peaks but with slightly different chemical shifts.

These interpolymers can be prepared by conducting the polymerization at temperatures of from about -30°C to about 250°C in the presence of such catalysts as those represented by the formula



wherein: each Cp is independently, each occurrence, a substituted cyclopentadienyl group π-bound to M; E is carbon or Si; M is a group IV metal, preferably Zr or Hf, most preferably Zr; each R is independently, each occurrence, hydrogen, hydrocarbyl, silahydrocarbyl, or hydrocarbylsilyl, containing up to about 30 preferably from 1 to about 20 more preferably from 1 to about 10 carbon or silicon atoms; each R' is independently, each occurrence, hydrogen, halo, hydrocarbyl, hyrocarbyloxy, silahydrocarbyl, hydrocarbylsilyl containing up to about 30, preferably from 1 to about 20, more preferably from 1 to about 10 carbon or silicon atoms or two R' groups together can be a C_1-C_{10} hydrocarbyl substituted 1,3-butadiene; M is 1 or 2; and optionally, but preferably in the presence of an activating cocatalyst. Particularly, suitable substituted cyclopentadienyl groups include those illustrated by the formula:



wherein each R is independently, each occurrence, hydrogen, hydrocarbyl, silahydrocarbyl, or hydrocarbylsilyl, containing up to about 30, preferably from 1 to about 20, more preferably from 1 to about 10 carbon or silicon atoms or two r groups together form a divalent derivative of such group. Preferably, R independently each occurrence is (including where appropriate all isomers) hydrogen, methyl, ethyl, propyl, butyl, pentyl, hexyl, benzyl, phenyl or silyl or (where appropriate) two such R groups are linked together forming a fused ring system such as indenyl, fluorenyl, tetrahydroindenyl, tetrahydrofluorenyl, or octahydrofluorenyl.

Particularly preferred catalysts include, for example, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl) zirconium dichloride, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl) zirconium 1,4-diphenyl-1,3-butadiene, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl) zirconium di-C_{1-4} alkyl, racemic-(dimethylsilanediyl)-bis-(2-methyl-4-phenylindenyl) zirconium di-C_1-C_4 alkoxide, or any combination thereof and the like.

It is also possible to use the following titanium-based constrained geometry catalysts, [N-(1,1-dimethylethyl)-1,1-dimethyl-1-[(1,2,3,4,5-η)-1,5,6,7-tetrahydro-s-indacen-1-yl]silanaminato(2-)-N]titanium dimethyl; (1-indenyl)(tert-butylamido)dimethyl-silane titanium dimethyl; ((3-tert-butyl)(1,2,3,4,5-η)-1-indenyl)(tert-butylamido) dimethylsilane titanium dimethyl; and ((3-iso-propyl)(1,2,3,4,5-η)-1-indenyl)(tert-butyl amido)dimethylsilane titanium dimethyl, or any combination thereof and the like.

Further preparative methods for the interpolymers used in the present invention have been described in the literature. Longo and Grassi (Makromol. Chem., Volume 191, pages 2387 to 2396 [1990]) and D'Anniello et al. (Journal of Applied Polymer Science, Volume 58, pages 1701-1706 [1995]) reported the use of a catalytic system based on methylalumoxane (MAO) and cyclopentadienyltitanium trichloride ($CpTiCl_3$) to prepare an ethylene-styrene copolymer. Xu and Lin (Polymer Preprints, Am. Chem. Soc., Div. Polym. Chem., Vol. 35, pages 686-687 [1994]) have reported copolymerization using a $MgCl_2/TiCl_4/NdCl_3/ Al(iBu)_3$ catalyst to give random copolymers of styrene and propylene.

Lu et al. (Journal of Applied Polymer Science, Vol. 53, pp. 1453 to 1460, 1994) have described the copolymerization of ethylene and styrene using a $TiCl_4/NdCl_3/ MgCl_2 /Al(Et)_3$ catalyst. Sernetz and Mulhaupt, (Macromol. Chem. Phys., Vol. 197, pp. 1071-1083, 1997) have described the influence of polymerization conditions on the copolymerization of styrene with ethylene using $Me_2Si(Me_4Cp)$(n-tert-butyl)$TiCl_2$/methylaluminoxane Ziegler-Natta catalysts. Copolymers of ethylene and styrene produced by bridged metallocene catalysts have been described by Arai, Toshiaki and Suzuki (Polymer Preprints, Am. Chem. Soc., Div. Polym. Chem., Volume 38, pages 349-350, 1997; U.S. Patent No. 5,883,213 and DE-A-197 11 339) and in U.S. Patent No. 5,652,315, issued to Mitsui Toatsu Chemicals, Inc.. The manufacture of α-olefin/vinyl aromatic monomer interpolymers such as propylene/styrene and butene/styrene is as described in US Patent No. 5,244,996, issued to Mitsui Petrochemical Industries Ltd.. All the above methods disclosed for preparing the interpolymer component are incorporated herein by reference. Also, the random copolymers of ethylene and styrene as disclosed in Polymer Preprints Vol. 39, No. 1, March 1998 by Toru Aria et al. can also be employed for the purposes of the present invention.

Polymerizations and unreacted monomer removal at temperature above the autopolymerization temperature of therespective monomers may result in formation of some amounts of homopolymer polymerization products resulting from free radical polymerization. For example, while preparing a substantially random interpolymer, an amount of atactic vinyl aromatic homopolymer may be formed due to homopolymerization of the vinyl aromatic monomer at elevated temperatures. The presence of vinyl aromatic homopolymer is in general not detrimental for the purposes of the present invention and can be tolerated. The vinyl aromatic homopolymer may be separated from the interpolymer, if desired, by extraction techniques such as selective precipitation from solution with a non-solvent for either the interpolymer or the vinyl aromatic homopolymer.

The substantially random interpolymer typically contains from 0.5 to about 65 mole percent , preferably from about 1 to about 55, more preferably from about 2 to about 50 mole percent of at least one vinyl or vinylidene aromatic monomer; or at least one hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer; or both; and from about 35 to 99.5 mole percent, preferably from about 45 to about 99, more preferably from about 50 to about 98 mole percent of ethylene and/or at least one aliphatic α-olefin having from about 3 to about 20 carbon atoms. The substantially random interpolymers typically contain from 0 to

about 20 mole percent of other polymerizable ethylenically unsaturated monomer or monomers.

The most preferred interpolymers for use as component (B) in the present invention are substantially random interpolymers comprising one or more vinyl aromatic monomers in combination with ethylene or a combination of ethylene and one or more $C_3 - C_8$ alpha olefin monomers, or a combination of ethylene and norbornene. Such interpolymers include the substantially random interpolymers selected from the group consisting of ethylene/styrene, ethylene/propylene/styrene, ethylene/butene/styrene, ethylene/pentene/styrene, ethylene/hexene/styrene, or ethylene/octene/styrene.

Those components (B) according to the present invention which comprise two or more interpolymers as defined above preferably comprise at least one such interpolymer which is substantially random.

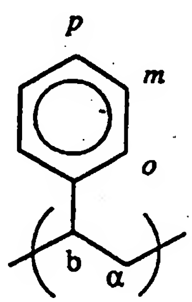
The melt index (MI) according to ASTM D-1238, Procedure A, condition E, generally is at least 0.01, typically from about 0.01 to about 50 g/10 minutes, preferably from about 0.01 to about 20 g/10 minutes, more preferably from about 0.1 to about 10 g/10 minutes, and most preferably from about 0.5 to about 5 g/10 minutes. The glass transition temperature (Tg) of the substantially random interpolymers is preferably from about -40°C to about +50°C, preferably from about 0°C to about +35°C, most preferably from about +10°C to about +30°C, measured using dynamic mechanical spectroscopy (DMS) techniques. The density of the substantially random interpolymer is generally about 0.930 g/cm^3 or more, preferably from about 0.930 to about 1.045 g/cm^3, more preferably from about 0.930 to about 1.040 g/cm^3, most preferably from about 0.930 to about 1.030 g/cm^3. The molecular weight distribution, M_w/M_n is generally from about 1.5 to about 20, preferably from about 1.8 to about 10, more preferably from about 2 to about 5.

The interpolymer styrene content and the content of atactic polystyrene (aPS) in an interpolymer of component (B) wherein the vinyl aromatic monomer is styrene can be determined using methods known in the art, in particular using proton nuclear magnetic resonance (^{1}H-NMR). The following protocol illustrates the determination of the interpolymer styrene content and the aPS content for an ethylene/styrene interpolymer and an ethylene/propylene/styrene interpolymer.

The interpolymer is dissolved in 1,1,2,2-tetrachloroethane-^{2}D (TCE-^{2}D) such that the resulting solution contains from about 1.6 to about 3.2 weight percent polymer. The

sample is heated in a water bath to soften the interpolymer. Proton NMR spectra are accumulated at 300 MHz or 400 MHz with the sample probe at a temperature in the range of from 80°C to 110°C, and referenced to the residual protons of TCE-^{2}D at 6 ppm. The following instrumental conditions are suitable for analysis of the interpolymer sample: standard ^{1}H; Sweep Width, 5000 Hz; Pulse Width, 90°; Recycle time 12 seconds.

Initially, a ^{1}H-NMR spectrum for a sample of polystyrene, Styron™ 680 Polystyrene (available from The Dow Chemical Company, Midland, Michigan, U.S.A.) is acquired. The protons are "labeled": b, branch; α, alpha; o, ortho; m, meta; p, para, as indicated in the following formula.



Integrals are measured around the protons labeled in the formula. Integral $A_{7.1}$ (aromatic, around 7.1 ppm) represents the three ortho/para protons; and integral $A_{6.6}$ (aromatic, around 6.6 ppm) the two meta protons. The two aliphatic protons labeled α resonate at 1.5 ppm; and the single proton labeled b is at 1.9 ppm. The aliphatic region is integrated from about 0.9 to 2.5 ppm and is referred to as A_{al}. The theoretical ratio for $A_{7.1}$: $A_{6.6}$: A_{al} is 3: 2: 3, or 1.5: 1: 1.5, and correlates very well with the observed ratios for the Styron™ 680 Polystyrene sample for several delay times of 1 second. The ratio calculations used to check the integration and verify peak assignments are performed by dividing the appropriate integral by the integral $A_{6.6}$. Ratio A_r is $A_{7.1} / A_{6.6}$.

Region $A_{6.6}$ is assigned the value of 1. Ratio A_l is integral $A_{al} / A_{6.6}$. All spectra collected have the expected 1.5: 1: 1.5 integration ratio of *(o+p): m:*(α+b). The ratio of aromatic to aliphatic protons is 5 to 3. An aliphatic ratio of 2 to 1 is predicted based on the protons labeled α and b, respectively, in the above formula. This ratio is also observed when the two aliphatic peaks are integrated separately.

For an ethylene/styrene interpolymer, the ^{1}H- NMR spectra have integrals $C_{7.1}$, $A_{6.6}$, and C_{al} defined such that the integration of the peak at 7.1 ppm includes all the aromatic protons of the copolymer as well as the ortho and para protons of aPS. Likewise, integration of the aliphatic region C_{al} in the spectrum of the interpolymers includes aliphatic protons from both the aPS and the interpolymer with no clear baseline resolved signal from either polymer. The integral of the peak at 6.6 ppm $A_{6.6}$ is resolved from the other aromatic signals and it is believed to be due solely to the aPS homopolymer (probably the *meta* protons). (The peak assignment for atactic polystyrene at 6.6 ppm (integral $A_{6.6}$) is made based upon comparison to the authentic sample of Styron™ 680 Polystyrene.) This is a reasonable assumption since, at very low levels of atactic polystyrene, only a very weak signal is observed here. Therefore, the phenyl protons of the copolymer should not contribute to this signal. With this assumption, integral $A_{6.6}$ becomes the basis for quantitatively determining the contents of aPS (atactic polystyrene).

For an ethylene/propylene/stryrene interpolymer, C_{al} also includes the protons of the propylene unit in the polymers backbone and an additional integral C_{Me} (0.9 to 0.5 ppm) is defined in the ^{1}H- NMR spectrum from the protons of the methyl side chain.

The following equations are then used to determine the degree of styrene incorporation in the ethylene/styrene interpolymer, and styrene and propylene in the ethylene/propylene/stryrene interpolymer, respectively:

$s_c = (C_{7.1} - (1.5 \text{ x } A_{6.6})) / 5$

$e_c = (C_{al} - C_{Me} - (1.5 \text{ x } A_{6.6}) - (3 \text{ x } s_c)) / 4$

$p_c = (C_{Me}) / 3$

$E = e_c / (e_c + s_c + p_c)$

$S_c = s_c / (e_c + s_c + p_c)$

$P = p_c / (e_c + s_c + p_c)$.

The following equations are used to calculate the mole percent ethylene, propylene and styrene in the interpolymers.

$$Wt\%E = \frac{E * 28}{(E * 28) + (S_c * 104) + (P * 42)}(100),$$

$$Wt\%S_c = \frac{S_c * 104}{(E * 28) + (S_c * 104) + (P * 42)}(100),$$

and

$$Wt\%P = \frac{P*42}{(E*28)+(S_c*104)+(P*42)}(100)$$

wherein : s_c, e_c and p_c are styrene, ethylene and propylene proton fractions in the interpolymer, respectively, and S_c ,E and P are mole fractions of styrene monomer, ethylene monomer and propylene monomer in the interpolymer, respectively.

The weight percent of aPS in the interpolymer can be determined using the following equation:

$$\mathrm{Wt\%aPS} = \frac{(\mathrm{Wt\%S})*\left(\frac{A_{6.6}/2}{S_c}\right)}{100+\left[(\mathrm{Wt\%S})*\left(\frac{A_{6.6}/2}{S_c}\right)\right]}*100$$

The total styrene content can also be determined by quantitative Fourier Transform Infrared Spectroscopy (FTIR).

The interpolymer of component (B) may be modified by typical grafting, hydrogenation, functionalizing, or other reactions well known to those skilled in the art. The polymer may be readily sulfonated or chlorinated to provide functionalized derivatives according to established techniques. The interpolymer may also be modified by various chain extending or cross-linking processes including, but not limited to peroxide-, silane-, sulfur-, radiation-, or azide-based cure systems. It may be advantageous to perform the cross-linking of the interpolymer component after the dielectric heating step, e.g. the HF welding step, said step involving one or more substrates comprising a composition of the present invention. A full description of the various cross-linking technologies is described in U.S. Patent No. 5,869,591 and EP-A-778,852, the entire contents of both of which are herein incorporated by reference. Dual cure systems, which use a combination of heat, moisture cure, and radiation steps, may be effectively employed. Dual cure systems are disclosed and claimed in EP-A-852,596, incorporated herein by reference. For instance, it may be desirable to employ peroxide crosslinking agents in conjunction with silane crosslinking agents, peroxide crosslinking agents in conjunction with radiation, and sulfur-containing crosslinking agents in conjunction with silane crosslinking agents. The interpolymer may also be modified by various cross-linking processes including, but not

limited to the incorporation of a diene component as a termonomer in its preparation and subsequent cross-linking by the aforementioned methods and further methods including vulcanization via the vinyl group using sulfur for example as the cross linking agent.

In addition to the above defined interpolymer component (B), the HF responsive compositions according to the present invention may optionally comprise one or more other polymers. Such polymer or polymers may prove especially useful in designing compositions and corresponding structures which have specifically desired or required performance attributes. Any other suitable polymer, for example an organic natural polymer or preferably an organic synthetic polymer, may be incorporated into the composition. One of skill in the art will choose a polymer that imparts certain desired characteristics to the composition of the invention and does not adversely affect its HF responsive properties. For example, incorporation of an additional polymers which is not an interpolymer within the definition of component (B) above may provide more preferred mechanical strength or tensile strength characteristics to a composition or structure according to the present invention. An additional advantageous result of incorporating another polymer in the interpolymer is economic in nature. The claimed compositions may be made increasingly cost efficient when the interpolymer is combined with a less expensive polymer that affords desirable characteristics.

The present invention expressly includes HF responsive compositions in which an additional polymer is blended with the interpolymer in amounts necessary to impart desirable qualities to the composition as a whole.

Suitable optional homopolymers and copolymers for blending with the above-defined interpolymers and incorporation in the compositions of the invention include , but are not limited to, homopolymers of ethylene or aliphatic alpha-olefins having from 3 to 8 carbon atoms, such as polyethylene or polypropylene, and copolymers of ethylene and one or more alpha-olefins having from 3 to 8 carbon atoms, such as ethylene-propylene copolymers, ethylene-1-octene copolymers, propylene-ethylene random interpolymers; interpolymers of propylene and at least one α-olefin containing from 4 to about 8 carbon atoms, terpolymers of ethylene, propylene and a diene, such as ethylene propylene diene rubber (EPDM) or rubber-toughened polypropylene. The olefinic polymer may also contain,

in addition to the α-olefin, one or more non-aromatic monomers interpolymerizable therewith, such as C_4-C_{20} dienes, preferably butadiene or 5- ethylidene-2-norbornene.

Suitable ethylene homo- and interpolymers include, for example, ethylene polymers, for example low density polyethylene (LDPE), high density polyethylene (HDPE), heterogeneous or homogeneous linear low density polyethylenes (LLDPE), such as ENGAGE™ polyolefin elastomers (POE) or Exact™ polymers, very- or ultra- low density polyethylenes (VLDPE or ULDPE), and substantially linear ethylene polymers, as disclosed e.g. in U.S. Patents Nos. 5,380,810; 5,272,236 and 5,278,272, or sold under the tradename AFFINITY™ by The Dow Chemical Company.

Suitable optional copolymers further include interpolymers containing one or more polar groups, preferably ethylene copolymers containing one or more polar groups, such as EVA, copolymers of ethylene and one or more alkyl acrylates including, for example, acrylic esters of linear, branched or cyclic alcohols of 1 to about 28 carbon atoms, such as methyl acrylate, ethyl acrylate, hydroxyethyl acrylate, and butyl acrylate, and a copolymer of ethylene and carbon monoxide.

Styrenic block polymers are also useful as an additional, optional polymer in the compositions according to the present invention. The term "block copolymer" is used herein to mean elastomers having at least one block segment of a hard polymer unit and at least one block segment of a rubber monomer unit. However, the term is not intended to include thermoelastic ethylene interpolymers which are, in general, random polymers. Preferred block copolymers contain hard segments of styrenic type polymers in combination with saturated or unsaturated rubber monomer segments. Suitable block copolymers having unsaturated rubber monomer units include, but are not limited to, styrene-butadiene (SB), styrene-isoprene (SI), styrene-butadiene-styrene block copolymers (SBS), styrene-isoprene-styrene (SIS), α-methylstyrene-butadiene-α-methylstyrene or α-methylstyrene-isoprene-α-methylstyrene, styrene-ethylene-butadiene-styrene (SEBS) and styrene acrylonitril (SAN) block polymer. The styrenic portion of the block copolymer is preferably a polymer or interpolymer of styrene and its analogs and homologs including α-methylstyrene and ring-substituted styrenes, particularly ring-methylated styrenes.

Preferred are optional polymers which blend well with the interpolymer(s) of component (B) and which do not require the addition of compatabilizer. The interpolymers for use as component (B) and in particular ethylene/styrene interpolymers are known to

exhibit compatibility with a wide range of polymers due to their olefinic and styrenic functionality. Examplary blends of ethylene/styrene interpolymers with other polymers and their benefits in certain structures and applications are described by C.F. Diehl et al., 57th SPE ANTEC (1999), pages 2149 - 2153, which publication is herein incorporated by reference in its entirety. The blends disclosed therein are also suitable for use in the compositions of the present invention. The optionally added polymer may be HF responsive itself and further enhance the HF responsiveness of the compositions according to the present invention. This is the case, for example, if the optionally added polymer is EVA.

The optional polymer is herein referred to as component (C) of the compositions according to the present invention. Component (C) may consist of one additional polymer, or comprise two or more polymers which are additional to and different from the interpolymer component(B). Preferably, the HF responsive composition of the present invention contains up to about 50 weight percent, preferably up to about about 30 weight percent of component (C) polymer or polymers.

Optionally, the HF responsive compositions of the present invention may also comprise a component (D) consisting of one or more inorganic fillers. Examples of suitable inorganic fillers are, for example, talc, calcium carbonate, aluminum trihydrate (ATH), glass fibers, marble dust, cement dust, magnesium oxide, magnesium hydroxide, antimony oxide, titanium dioxide, or chalk, including mixtures thereof. Preferred inorganic fillers are calcium carbonate and ATH. As compared to structures and articles made from a corresponding composition without such filler or fillers, the incorporation of filler allows, for example, for structures and articles with improved properties, such as improved hardness and tensile modulus while generally maintaining good elongation properties, a better scratch resistance and/or flame retardant properties, or for reducing the costs of the final composition without compromising on the desired properties. The definition of component (D) excludes any molecular sieve material suitable as component (A).

The amount of inorganic filler incorporated in the HF responsive composition of the invention largely depends upon the desired end use of said composition. Generally, the maximum acceptable amount of filler depends upon the density of the inorganic filler. The higher the filler density, the higher the maximum acceptable amount of filler in the composition (based on the total weight of the composition). Preferably, the amount of

component (D) in the compositions of the invention ranges from 0 to about 70 weight percent (based on the total weight of components (A), (B), (C) and (D) in the composition). Advantageously, the combined amounts of the molecular sieve component (A) and filler component (D) do not exceed about 70 weight percent. The incorporation of filler allows, for example, for structures and articles with improved properties, as compared to structures and articles made from a corresponding composition lacking a filler, such as, for example, improved hardness and tensile modulus while good elongation properties are maintained, a better scratch resistance and/or flame retardant properties, or for reducing the costs of the final composition without compromising on the desired properties. Filled polymer compositions comprising a substantially random interpolymer within the definition of component (B) and one or more inorganic fillers are disclosed in U.S. Patent No. 5,973,049 by Bieser et al., which is herein incorporated by reference in its entirety.

The HF responsive composition of the present invention may also include one or more conventional additives which impart a desirable functional property to the composition or structure of the invention, but neither substantially detract from HF responsiveness of the composition, nor otherwise adversely affect its desired combination of properties. Such additives include, without limitation, antioxidants, such as hindered phenols or phosphites, or process stabilizers, lubricants, light and ultraviolet (UV) stabilizers, such as hindered amines, plasticizers, such dioctylphtalate or epoxidized soy bean oil, tackifiers, such as known hydrocarbon tackifiers, waxes, such as polyethylene waxes, processing aids, such as oils, talc, stearic acid or a metal salt thereof, crosslinking agents, such as peroxides or silanes, colorants or pigments, blowing agents and biocides - in an amount which does not interfere with the desired properties of the HF responsive composition of the present invention. The additives are employed in functionally suitable amounts known to those skilled in the art, for example generally in amounts of up to about 10, preferably in amounts of from about 0.01 to about 5 percent, more preferably in amounts of from about 0.02 to about 2 percent, based on the total weight of the HF responsive composition.

In the HF responsive compositions according to the present invention, the proportions of the molecular sieve component (A) and the interpolymer component (B) may

be varied within a relatively wide range. For example, as little as about 2 % by weight of component (A) added to about 98 % by weight of component (B) (based on the total of weight of components (A), (B), (C) and (D)) has been found to exert a desirable enhancing effect on the dielectric properties of the composition. Typically, the interpolymers used as component (B) are not HF responsive as such. HF responsiveness of the compositions acccording to the present invention is found to correlate with the amount of molecular sieve material comprised therein. Higher amounts of molecular sieve provide for increased HF responsiveness of the compositions. The amount of molecular sieve comprised in the composition of the invention should be high enough to allow efficient dielectric heating of the composition, in particular efficient HF welding of a structure or article comprising the composition of the invention. The improvement provided by the present invention comprises blending a molecular sieve material (A) with the interpolymer component (B) which component (B) as such does not respond, or which only insufficiently responds to dielectric heating, and optionally additional components (C) and (D) and subsequently subjecting the blend to a dielectric heating process.

Compositions comprising an optional component, which is HF active, for example EVA, are also within the scope of the present invention. In such compositions, component (A) is used to significantly enhance HF responsiveness, as compared to the corresponding composition lacking component (A). Advantageously, the compositions according to the present invention comprise about 2 weight percent or more, preferably about 4 weight percent or more of the molecular sieve component (A), for example zeolite, based on the combined weights of components (A) to (D) present in the composition. Up to about 50 % by weight or more of component (A) may be incorporated whereby the dielectric properties of the resulting product improve accordingly. More than 50 % of component (A) may also be incorporated in the composition of the invention, but such tends to change the properties of the resulting product. Typically ,the compositions of the invention contain less than about 50 weight percent of the molecular sieve component (A), preferably less than about 30 weight percent (based on the combined weights of components (A) to (D) present in the composition).

The amount of the interpolymer component (B) in the compositions of the present invention may range from at least about 30 weight percent or more to about about 98 weight percent or less, preferably 96 weight percent or less (based on the combined weights of

components (A) and (B) in the composition). Regarding practical and/or commercial viability of the present invention, incorporation of from about 2 % to about 30 % by weight of the molecular sieve component (A) has been found preferable.

In a preferred embodiment, the present invention provides a HF responsive composition comprising:

(A) from about 2 to about 50 weight percent of a molecular sieve material,

(B) from about 98 to about 30 weight percent of at least one interpolymer comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c),

(C) from 0 to about 50 weight percent of a polymer which is not an interpolymer defined as component (B), and

(D) from 0 to about 70 weight percent of an inorganic filler which is not a molecular sieve material defined as component (A).

These weight percentages are based on the total amounts of components (A), (B), (C) and (D) present in the composition.

Preferred are HF responsive compositions comprising the components (A), (B), (C) and/or (D), which are indicated herein as preferred, more preferred or particularly preferred, and any combination of such components.

Preferably, the polymer materials used in the composition of the present invention are halogen-free, meaning that the polymer materials lack chemically combined halogen atoms. In other words, halogenated monomers do not constitute building blocks for such

preferred polymer materials and the polymers are not halogenated subsequent to formation. The compositions according to the invention may, however, contain small amounts of non-polymeric halogenated additives, including conventional halogenated flame retardant additives.

If desired or required, the compositions according to the present invention are selected such that they avoid or reduce the environmental concerns which have been raised for PVC products. Such selection is within the routine skills of the person skilled in the art.

One aspect of the present invention relates to a process for preparing a composition of the present invention. The process comprises blending the molecular sieve component (A), the interpolymer component (B) and optionally further components of the HF responsive composition of the present invention. The process comprises the essential steps of blending from about 2 percent to about 50 percent by weight or more of a molecular sieve material (A) with at least one interpolymer within the definition of component (B) to obtain a uniform dispersion of the molecular sieve material throughout the interpolymer and subjecting the resulting blend to dielectric heating, preferably by passing the blend through a high frequency alternating electrical field. The blend is delivered and passed through the high frequency electrical field by any one of several conventional means, for example, by a conveyor belt for carrying the blend through the electrical field.

Alternatively, the process may comprise the steps of blending from about 2 percent to about 50 percent by weight of the molecular sieve component (A) with the interpolymer component (B), and optional additional components to obtain a uniform dispersion of (A) throughout (B), forming the blend into a structure or a shaped article and subjecting the structure or the shaped article to a high frequency, alternating electrical field thereby producing a weld involving the composition of the invention.

The composition according to the present invention can be prepared by any conventional and appropriate compounding operation. Preparation of the composition may involve, for example, physical admixture, including, for example, dry blending and solution blending. If appropriate, polymer blends may be formed *in-situ* and subsequently mixed with the molecular sieve material and any desired optional component. The interpolymer and the molecular sieve are preferably used in the form of pellets or dry powder. In such form, they can easily be dry blended in any required proportions by weight and be fed into the extrusion, spinning, or molding device, and formed into structures or fabricated articles.

The compositions of the invention are HF responsive, meaning that the compositions can be heated rapidly and efficiently by application of a time varying electric field generated at frequencies of 0.1 – 30,000 MHz to generate heat inside the composition. The HF range generally covers three frequency ranges commonly referred to as the ultrasonic frequency range (18 kilohertz (kHz) – 1 MHz), the RF (radio frequency) range (1MHz – 300 MHz), and the MW (microwave) frequency range (300 MHz – 10,000 MHz). The RF and MW ranges are of particular interest.

Generally, dielectric heating can be performed at a wide variety of frequencies. However, to ensure that there is no interference with communication services, there are regulatory restrictions on the frequencies which can be used. The permitted frequencies differ from country to country and include the following ranges

13.56 MHz ± 0.06 %
27.12 MHz ± 0.6 %
40.68 MHz ± 0.05 %
433.92 MHz ± 0.2 %
896/915 MHz ± 10/13 MHz
2450 MHz ± 50 MHz
5800 MHz ± 75 MHz
22125 MHz ± 125 MHz

HF-responsiveness of the compositions is determined by their dielectric loss properties, as reflected, for example, in the dielectric loss factor (DLF). The higher the dielectric loss factor of the composition of the invention, the more efficient is its response to dielectric heating. The dielectric loss tangent (tan δ), also referred to as dissipation factor (DF), calculates as the dividend of a material's DLF and the dielectric constant (DC). The DLF and DC are dependent on the frequency and the temperature.

It has been found that compositions according to the invention with a dielectric loss factor (DLF) of even below 0.01 can be HF welded. Preferred are compositions having a DLF of 0.01 or higher, more preferably of 0.05 or higher, as measured in the frequency range of from about 13 to about 40 MHz and from about ambient temperature to about 80°C. Advantageously, the DLF of a HF weldable composition of the present invention is selected such that the HF welding step can be performed with appropriate efficiency, e.g.

under high speed manufacturing conditions, in particular conditions used for commercial production. The quality of the weld, reflected e.g. by tightness, reliability, peelability or seal strength, is selected such that the weld meets the requirements of the application.

The DC and DLF are readily determined by known instrumented dielectric testing methods. A preferred test fixture uses a Hewlett-Packard Impedance/Material Analyzer, Model 4291 B coupled with a a Hewlett-Packard Dielectric Test Fixture, Model 16453A. Dielectric properties can be measured according to ASTM D-150 on compression molded plaques, or preferably on film strips, formed from the material to be analyzed, e.g. a composition according to the present invention.

The essence of HF techniques is that an HF voltage is applied across the plates of a capacitor to give an alternating electric field. The workpiece is held or passed between the plates and heated. Size and shape of the capacitor can be varied to suit the particular process subject to certain constraints in HF energy generation. The HF output available from typical generators is at least several kilowatt (kW). Efficiency of energy utilization is about 50 % or more. The heat-flow rate is mainly determined by the desired production rate. For a uniform electric field, the amount of power (P) adsorbed by a material exposed to said field is given by the equation:

$$P = 2\pi f\, CE^2 \varepsilon \tan \delta,$$

wherein P is the power or energy per time absorbed by the material in watts (W = Joule per second), f is the frequency (MHz), E is the field strength, or voltage (gradient) between the electrodes in kilovolt (kV) generated by the HF generator, C is the capacitance of the material in picofarads (pF), ε is the dielectric constant of the material and $\tan \delta$ is the dielectric loss tangent or dissipation factor (DF). The time needed to attain the temperature required to plastify or melt the polymer or polymers comprised in the composition of the invention is less at higher field strength and higher frequency. Under commercial circumstances, it may be desirable to choose the conditions such that the heating phase is relatively shorter. Practically, the frequency is maintained at HF frequencies allocated by the national authority for industrial HF heating and fixed for a given equipment. In general, the maximum field strength (E) varies with the system and apparatus used. For a specific equipment, the maximum field strength can be varied with the power setting which may be set to range from 0 to 100 (in scale units).

The HF responsive compositions of the invention allow processing by means of HF radiation. In operations, for example drying, preheating, curing, foaming, shaping, injection molding, transfer molding, blow molding, flow molding, softening and HF welding, the compositions can be exposed to an alternating high frequency field to provide the needed heat to accomplish the desired operation.

For example, the present invention provides an efficient means for quickly preheating a composition of the invention before processing it, for example, before extruding, injection or compression molding it and the like. The compositions of the invention are also suitable for use in HF extruders, including continuous extruders or batch extruders. For example, wire and cable coating comprising a composition according to the present invention can be applied in a continuous HF extruder.

Preferred according to the present invention is an HF responsive composition which allows for the formation of a HF weld involving such composition. Such HF weldable composition, or rather a structure or fabricated article comprising such composition, can be bonded to itself or to another material using HF heating and MW heating rather than conventional heat sealing. In the context of HF techniques, the terms "sealing" and "welding" (and variations of each term) are used interchangeably herein. In order to attain the best possible seal it is highly desirable to attain a uniform generation and distribution of heat in the welding area. Hence, the composition of the invention should be substantially homogeneous, the molecular sieve material and the interpolymer component (B) being uniformly distributed, at least in any area or location intended to participate in weld formation. The HF welding technique is advantageously used e.g. in the mass production of fabricated plastic articles which require a reliable connection between different plastic materials, including, for example, structural integration of a supplementing part in a principal part. It may be particularly advantageous to use the same material for the parts to be affixed to each other as that can reduce costs. The utilization of the HF welding technique increases reliability and production yield.

Known HF welding techniques include welding, tear-seal welding and appliqué welding. Plain welding is the welding of two or more thicknesses of material. Optionally, the welding tool may be engraved to provide the welded area with a decorative appearance. Tear seal welding involves simultaneous sealing and cutting. Appliqué welding is a

technique where a piece of material is cut out and HF welded to the surface of another to provide an ornamental effect.

For the purpose of the present invention, the quality of the HF weld is such that it serves the purpose of the article of manufacture. Depending on the intended application, it may be desirable to form a HF weld which can be peeled apart or separated on application of a certain force without rupturing the materials forming the weld, or a tight and reliable weld which cannot be separated without rupturing the materials forming the weld. The formation of a HF weld requires that the thermoplastic polymer forming the weld is HF heated to a high enough temperature which enables molecular entanglement and formation of an integral connection. This requires not only softening but melting of the polymers forming the weld, however, without decomposition. Typically, pressure is applied to support the bonding and integral connection .

HF welding technology including suitable machinery and the variable process parameters are well known in the art. Many different types of welding machines are available to allow for production of a wide range of diverse fabricated articles and the diverse applications. The selection including optimization of a HF welding technique, suitable HF welding machinery and advantageous HF welding process parameters for the production of a structure or fabricated article according to the present invention is routine for the person of ordinary skill in the art. Major factors impacting said choices include, for example, type and dimension of the structure or fabricated article and cost effiency of the production process.

Typically, HF welding machines comprise at least a HF power generator, a press, a workpiece handling mechanism and a control system. In Europe, the generator usually operates at a frequency of 27.12 MHz (± 0.6 %). The maximum output power level, (measured in Watts or kilowatts), is a machine dependent parameter, which is also referred to as the machine rating. The efficiency of a HF generator typically is in the range of from 50 - 60 percent, meaning about 50 - 60 percent of its input power is converted into HF power. The HF power required to weld a given area depends upon several factors including, for example, the dimension of the area and the type of material to be welded, thickness of material, the type and design of the electrode and desired welding time. The output power level can be adjusted to suit the conditions required for the weld. The press provides the means for pressing welding tools against work materials during HF power input, that is the

heating phase of the welding cycle, and – to the extent desired or required - during the cooling phase. The handling mechanism feeds the workpiece to the press, positions it under the press for welding and removes the work piece after welding. Available types of handling mechanisms include manual, linear, rotary and rail mechanisms. The control system serves to ensure that the machine can be operated efficiently with good repeatablility and safely. Control systems range from simple mechanical and electrical controls on small machines to full machine and process automation on large complex machines.

The process parameters should be selected and controlled such that a weld meeting the required quality standard is obtained, advantageously under conditions suitable for large scale production. Controllable process parameters include, for example, stroke and pressure of the welding press, HF output power and the timing of the heating and cooling cycles. HF power and welding time should be adjusted such to avoid over-heating and possible negative effects on the workpiece or the tools.

Detailed teachings concerning the welding process and suitable welding equipment are seen, for example, in the "High Frequency Welding Handbook" published by The Federation of High Frequency Welders, 66 East Street, Ashburton, Devon TQ 13 7AX, United Kingdom, in particular in Chapter 2 entitled "The HF Welding Process", which is incorporated herein by reference.

The present invention further provides HF compositions which have undergone a film-forming, fiber-forming, foam-forming, profile-forming, coating or molding process. Such compositions are collectively referred to as structures according to the invention. Such structures in the form of a film (including thick film or sheet), fiber, foam, profile or molding are obtainable using a single process step and may be monolayer or multilayer, e.g. two-, three-, four-, or five-layer structures. Multilayer structures may comprise one or more layers which are formed from materials other than a composition of the invention provided that they are made in a single process step. The structures according to the invention may be assembled into articles of manufacture in one or more additional process steps. Such articles of manufacture are also within the scope of the present invention.

In particular, the present invention provides HF compositions in the form of a film. A film structure according to the present invention comprises one or more layers and is made in a single process step employing a HF responsive composition according to the

invention, e.g. by co-extrusion or extrusion coating. Films comprising a HF responsive composition according to the invention which are made in a multiple step process (two or more steps) are considered herein as articles of manufacture and are also within the scope of the present invention.

The present invention also relates to foamed compositions comprising components (A) and (B), and optionally components (C) and (D) and provides an improved process for preparing such foamed compositions. Such process comprises the heating of a composition of the invention comprising a conventional blowing agent by dielectric means. The heating rate of the polymer composition is significantly increased by incorporating a molecular sieve material (A) into the interpolymer component (B) either before, after or during incorporation of the conventional blowing agent into the polymer. Suitable conventional blowing agents include gases and volatile liquids at ordinary temperatures and pressures.

In yet another aspect, the present invention relates to molded compositions comprising components (A) and (B), and optionally components (C) and (D).

The compositions according to the invention are formed into structures and articles of manufacture. Examples include such structures and fabricated articles which currently rely on HF welds involving flexible PVC or PVCD polymers, and wherein these polymers can suitably be replaced with a composition according to the present invention. The advantages provided by the HF responsive compositions according to the present invention include faster and more efficient sealability, sealing through poor heat conductors, e.g. paper or cardboard exteriors, stronger seals, the ability to seal, bond or laminate larger surface areas, sealing of thicker or more complex film laminates and specific sealing. In particular, HF sealing and bonding offers a performance advantage over conventional thermal or heat sealing when rapid sealing time is a crucial factor, such as in high speed manufacturing. HF sealing technologies allow energy to be concentrated at the HF active layer, thus eliminating the need to transfer heat through an entire structure or article. This is particularly advantageous for thick film or sheet (having a gauge of more than 125 microns) where conventional thermal sealing requires relatively long contact times to permit thermal transfer through the thick polymer structure to the bonding interface. HF sealing is also advantageous when a thermally sensitive material is used within the structure of article, such as a color sensitive dyed fabric or nonwoven, or an oriented film which can soften and

undesirably shrink upon heating. The HF responsive compositions can also be fabricated into very complex shapes, which is difficult to do with thermal sealing equipment. Furthermore, the compositions of the present invention enable structures, e.g. films, and fabricated articles, which have a good balance of mechanical properties and/or good haptics. The HF responsive compositions according to the present invention also allow the introduction of a pattern or a figure into a structure, such as a film, or a fabricated article provided herein. For example, such structure or fabricated article may be branded with a trademark.

For HF weldability of a multilayer structure or article of manufacture according to the present invention to itself or to another substrate, it is advantageous that the layer comprising the HF responsive composition of the invention is a surface layer or that it is in sufficiently close proximity to the surface bonding such that the heat generated by the HF activation of the layer comprising the composition according to the invention can quickly transmit to and through the surface bonding and cause thermal bonding and sealing.

The composition according to the present invention may be melted and formed into the molded article of manufacture by molding. Such articles are useful e.g. in automotive applications such as sun visors, trunk covers, door panels and dashboards.

The present invention also provides a fabricated article comprising a composition according to the invention. Preferably, the HF responsive composition of the invention confers HF sealability to the fabricated article of the invention, or a part thereof, and is designed to meet the needs of the application.The fabricated article can be made from one or more structures of the invention, including a combination of different structures, e.g. a foam and a film, or a fabric, e.g. a woven fabric, and a film, or directly from the compositions of the invention. Preferred are such fabricated articles which are produced or assembled using HF welding technology. More preferred are such fabricated articles comprising one or more seals formed from at least one composition according to the present invention using HF welding technology. In another embodiment, such seal may be formed from at least one composition of the present invention and another HF weldable material, advantageously a HF weldable thermoplastic material. Fabricated articles according to the invention which can be produced using HF welding include, for example, stationery, such as book covers, clipboards, labels, binders, notebooks, stationary wallets, ziploc bags and office files;

inflatable items, such as advertising items, e.g. items advertising novelties, toys, e.g. beach balls, air beds, water beds, rafts and life jackets; household items, such as upholstery, e.g. chair upholstery, headboards, quilting and table mats; everyday items, such as badges, key fobs, umbrellas or chequebook covers; large items, such as tarpaulins to protect and/or cover exposed objects, e.g. for trucks, boats, trains or containers, sign and banners, tents and marquees, liners, e.g. pool liners;; medical devices, such as containers for storing medical solutions or blood products, ostomy bags or blood bags. The fabricated articles of the invention may be made from multilayer structures, in particular multi-layer films, or coated fabrics. Layers or plies of the compositions according to the invention can be used as a means for sealing or bonding materials which, as such, cannot efficiently be HF sealed. Substrates, for example, particles, films, sheets, blocks, rods and spheres can be coated, at least in the area to be bonded by dielectric heating.

The structures and fabricated articles according to the present invention can be used for packaging applications, specifically in packaging applications where high speed seals are required, including, for example, pouches, bags including bags with a resealing means, e.g. a zipper or "zip-lock" type means, such as sandwich bags; laminates of foam, fabric or film layers and powder moldings. The structures and fabricated articles according to the invention can also be used for microwave sealing applications, such as microwave sealable containers and plastic bags, tube sealing, e.g. toothpaste and shampoo tube sealing, and valve bag sealing. The structures and fabricated articles according to the invention can also be used in automotive applications.

In a preferred aspect, the present invention relates to an article of manufacture fabricated from a monolayer or multilayer film structure of the invention. Examplary articles include, for example, coated substrates, such as coated fabrics and laminates. Such fabricated articles desirably include at least one segment wherein the film is sealed to itself, to a substrate, or to both, at a seal interface. Sealing preferably results from exposure of the film to HF energy. The seal interface preferably has a bond strength of at least 0.2 Newton per millimeter (N/mm), as determined by the seal strength test or by visual observation of the formation of the seal. Typically, such seal is still peelable apart. The films of the invention may be of any gauge that serves a given need, but typically have an overall gauge within a range of from 12 to 2500 micron, preferably from 25 to 1000 micron and most preferably from 25 to 1000 microns. Particularly preferred articles of manufacture include

coated fabrics comprising a fabric, e.g. a woven fabric, such as a woven polyester fabric, which is at least partically coated on at least one side with a structure according to the present invention, preferably a film. Representative examples of such coated fabrics are tarpaulins and artificial leather. Advantageously, the coated fabrics are produced via a calandering process.

Films of the invention (including film structures and articles of manufacture comprising such structure) which can replace flexible PVC and which are HF sealable, are suitable, for example, for applications in which the film, or the products in contact with the film, come into intimate contact with the human body. Such applications may include medical or urological collection bags, medical ostomy bags, medical infusion or intravenous (IV) bags, inflatable devices such as air mattresses, flotation devices or toys, food packaging, children's articles and toys, curtains, labels, reinforced laminates for tents and tarpaulins, roofing membranes, geomembranes and geotextiles, and stationery applications, such as notebook covers. Compositions according to the invention can also be extruded into a tubing with an HF responsive outer layer. Such tubing can readily be used in conjunction with HF weldable films to provide a complete HF weldable film structure such as a medical collection bag. The aforementioned listing is merely illustrative and can readily be expanded by the skilled person to virtually any device or application which requires and HF sealable, flexible monolayer or multilayer film, including a laminate or composite.

Any conventional film forming process may be used to fabricate films of the present invention. Illustrative processes suitable for use in making a monolayer or multilayer film according to the present invention include known processes to make calendered film, blown film or cast film as well as extrusion coating of one or more layers upon a film or substrate, or lamination of multiple plies of film layers. In addition, the films of the present invention can be fabricated into extruded profile shapes such as tubing.

Examples:

The following Examples are illustrative of the invention, but are not to be construed as limiting the scope thereof in any manner.

The following abbreviations are used in the Examples: ESI(s) mean substantially random ethylene-styrene interpolymer(s); EPS means substantially random ethylene-propylene-styrene interpolymer; CAS means Chemical Abstracts Registry; DC means

dielectric constant; DLF means dielectric loss factor; MI means melt index determined according to ASTM D-1238, condition E (2.16 kg, 190°C, for polypropylene 230°); n.a. means 'not analyzed'. The designations of components (A) – (D) are used as defined hereinabove.

Example 1

I) Preparation of Interpolymers (Component (B))

ESIs 1-4 and EPS-1 are prepared in a continuously operating loop reactor. An Ingersoll-Dresser twin screw pump provides the mixing. The reactor runs liquid full at 475 psig (3,275 kPa). Raw materials and catalyst/cocatalyst flows are fed into the reactor through injectors and Kenics static mixers in the loop reactor piping. From the discharge of the loop pump, the process flow goes through two shell and tube heat exchangers before returning to the suction of the loop pump. Upon exiting the last exchanger, loop flow returns through the injectors and static mixers to the suction of the pump. A second monomer/feed injector and mixer are used if available. Heat transfer oil or tempered water is circulated through the exchangers' jacket to control the loop temperature. The exit stream of the loop reactor is taken off between the two exchangers. The flow and solution density of the exit stream is measured by a Micro-Motion™ mass flow meter.

Solvent is injected to the reactor primarily as part of the feed flow to keep the ethylene in solution. A split stream from the pressurization pumps prior to ethylene injection is taken to provide a flush flow for the loop reactor pump seals. Additional solvent is added as a diluent for the catalyst. Feed solvent is mixed with uninhibited styrene monomer on the suction side of the pressurization pump. The pressurization pump supplies solvent and styrene to the reactor at approximately 650 psig (4,583 kPa). Fresh styrene flow is measured by a Micro-Motion™ mass flow meter, and total solvent/styrene flow is measured by a separate Micro-Motion™ mass flow meter. Ethylene is supplied to the reactor at approximately 690 psig (4,865 kPa). The ethylene stream is measured by a Micro-Motion™ mass flow meter. A flow meter /controller is used to deliver hydrogen into the ethylene stream at the outlet of the ethylene control valve. Propylene can be added either as a high pressure stream after the solvent pressurization pump, or upstream of the pump near the styrene injection.

The ethylene/hydrogen mixture is at ambient temperature when it is combined with the solvent/styrene stream. The temperature of the entire feed stream as it entered the reactor loop is lowered to approximately 2°C by a glycol cooled exchanger.

The catalyst system is a three component system composed of a titanium catalyst, an aluminum co-catalyst and a boron co-catalyst. Preparation of the three catalyst components takes place in three separate tanks. The titanium catalyst is (1H-cyclopenta[1]phenanthrene-2-yl)dimethyl(t-butylamido)-silanetitanium 1,4diphenylbutadiene) which is prepared as described under II), below. The aluminum co-catalyst component is a modified methylaluminoxane type 3A (MMAO-3A; CAS No. 146905-79-5) and the boron co-catalyst is tris(pentafluorophenyl)borane (FAB, CAS No. 001109-15-5). The molar ratios of boron co-catalyst to titanium catalyst (B/Ti) and aluminum co-catalyst to titanium catalyst (Al/Ti) which are employed to prepare the various individual interpolymers are listed in Table 1.

Table 1: Catalyst Molar Ratios used in the preparation of ESI 1-4 and EPS-1

Polymer	B/Ti molar ratio	Al/Ti molar ratio
ESI-1	4	7
ESI-2	4	5
ESI-3	4	5
ESI-4	4	8
EPS-1	4	5

Fresh solvent and concentrated catalyst/co-catalyst/secondary co-catalyst premix are added and mixed into their respective run tanks and fed into the reactor via a variable speed Pulsafeeder™ diaphragm pumps. As previously explained, the three component catalyst system enters the reactor loop through an injector and static mixer into the suction side of the twin screw pump. The raw material feed stream is also fed into the reactor loop through an injector and static mixer upstream of the catalyst injection point or through a feed injector/mixer between the two exchangers.

Polymerization is stopped with the addition of catalyst kill (water) into the reactor product line after the Micro-Motion™ mass flow meter measuring the solution density. A static mixer in the line provided dispersion of the catalyst kill and additives in the reactor effluent stream. This stream next enters post reactor heaters that provide additional energy

for the solvent removal flash. This flash occurrs as the effluent exits the post reactor heater and the pressure is dropped from 475 psig (3,275 kPa) down to approximately 450 mmHg (60 kPa) of absolute pressure at the reactor pressure control valve.

This flashed polymer enters the devolatilization section of the process. The volatiles flashing from the devolatilization are condensed with a glycol jacketed exchanger, passed through vacuum pump, and are discharged to vapor/liquid separation vessel. In the first stage vacuum system, solvent/styrene are removed from the bottom of this vessel as recycle solvent while unreacted ethylene exhausts from the top. The ethylene stream is measured with a Micro-Motion™ mass flow meter. The measurement of vented ethylene plus a calculation of the dissolved gases in the solvent/styrene stream are used to calculate the ethylene conversion. The polymer and remaining solvent are pumped with a gear pump to a final devolatilizer. The pressure in the second devolatilizer is operated at approximately 10 mmHg (1.4 kPa) absolute pressure to flash the remaining solvent. The dry polymer (< 1000 ppm total volatiles) is pumped with a gear pump to an underwater pelletizer, spin-dried, and collected.

The process conditions and amounts of monomers used to prepare the individual ethylene styrene interpolymers are summarized in Table 2.

Table 2: Process Conditions for Preparation of ESIs 1-4 and EPS-1

Inter-polymer	Reactor Temp.	Solvent Flow	Ethylene Flow	Propylene Flow	Hydrogen Flow	Styrene Flow	Ethylene Conversion
	°C	kg/h	kg/h	kg/h	kg/h	kg/h	%
ESI-1	110	9267	1369	0	0.066	660	89.8
ESI-2	113	10497	1277	0	0.015	976	90.7
ESI-3	83	7323	588	0	0	1799	92.1
ESI-4	57	143	15	0	0.0005	199	98.0
EPS-1	115	8543	1133	237	0	296	88.8

Table 3 lists certain properties characterizing the interpolymers used in the Examples. Interpolymer styrene content, interpolymer propylene content and content of atactic polystyrene are determined using the proton nuclear magnetic resonance method described hereinbefore.

Table 3: Properties of ESI-1-4 and EPS-1

Inter-polymer	Interpolymer Styrene	Interpolymer Propylene	Atactic Polystyrene	Melt Index
	weight %	weight %	weight %	g/10 min
ESI-1	30.1	0	0.6	0.93
ESI-2	38.5	0	1.2	0.76
ESI-3	68.9	0	6.7	1.10
ESI-4	77.4	0	7.6	0.74
EPS-1	14.1	16.5	0.1	1.23

II) Preparation of the Titanium Catalyst

1) Preparation of lithium 1H-cyclopenta[l]phenanthrene-2-yl

To a 250 mL round-bottom flask containing 1.42 g (0.00657 mole) of 1H-cyclopenta[l]phenanthrene and 120 mL of benzene is added dropwise 4.2 mL of a 1.60 M solution of n-butyllithium in mixed hexanes. The solution is allowed to stir overnight. The lithium salt is isolated by filtration, washed twice with 25 mL benzene and dried under vacuum. ^{1}H-NMR analysis indicates the predominant isomer is substituted at the 2 position.

2) Preparation of (1H-cyclopenta[l]phenanthrene-2-yl)dimethylchlorosilane

To a 500 mL round bottom flask containing 4.16 g (0.0322 mole) of dimethyldichlorosilane (Me_2SiCl_2) and 250 mL of tetrahydrofuran (THF) is added dropwise a solution of 1.45 g (0.0064 mole) of lithium 1H-cyclopenta[l]phenanthrene-2-yl in THF. The solution is stirred for approximately 16 hours, after which the solvent is removed under reduced pressure, leaving an oily solid which was extracted with toluene, filtered through a diatomaceous earth filter aid (Celite™), washed twice with toluene and dried under reduced pressure.

3) Preparation of (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamino)silane

To a 500 mL round-bottom flask containing 1.98 g (0.0064 mole) of (1H-cyclopenta[l]phenanthrene-2-yl)dimethylchlorosilane and 250 mL of hexane is added 2.00 mL (0.0160 mole) of t-butylamine. The reaction mixture is allowed to stir for several days, then filtered using a diatomaceous earth filter aid (Celite™) and washed twice with hexane. The product is isolated by removing residual solvent under reduced pressure.

4) Preparation of dilithio (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamido)silane

To a 250 mL round-bottom flask containing 1.03 g (0.0030 mole) of (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamino)silane) and 120 mL of benzene is added dropwise 3.90 mL of a solution of 1.6 M n-butyllithium in mixed hexanes. The reaction mixture is stirred for approximately 16 hours. The product is isolated by filtration, washed twice with benzene and dried under reduced pressure.

5) Preparation of (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium dichloride

To a 250 mL round-bottom flask containing 1.17 g (0.0030 mole) of $TiCl_3$•3THF and about 120 mL of THF is added at a fast drip rate about 50 mL of a THF solution of 1.08 g of dilithio (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamido)silane. The mixture is stirred at about 20°C for 1.5 hours at which time 0.55 grams (0.002 mole) of solid $PbCl_2$ is added. After stirring for an additional 1.5 h the THF is removed under vacuum and the residue is extracted with toluene, filtered and dried under reduced pressure to give an orange solid.

6) Preparation of (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium 1,4-diphenylbutadiene

To a slurry of (1H-cyclopenta[l]phenanthrene-2-yl)dimethyl(t-butylamido)-silanetitanium dichloride (3.48 g, 0.0075 mole) and 1.551 grams (0.0075 mole) of 1,4-diphenylbutadiene in about 80 mL of toluene at 70°C is added 9.9 mL of a 1.6 M solution of n-BuLi (0.0150 mole). The solution immediately darkens. The temperature is increased to bring the mixture to reflux and the mixture is maintained at that temperature for 2 hours. The mixture is cooled to about -20°C and the volatiles are removed under reduced pressure. The residue is slurried in 60 mL of mixed hexanes at about 20°C for approximately 16 hours. The mixture is cooled to about -25°C for about 1 hour. The solids are collected on a glass frit by vacuum filtration and dried under reduced pressure. The dried solid is placed in a glass fiber thimble and solid extracted continuously with hexanes using a soxhlet extractor. After 6 hours a crystalline solid is observed in the boiling pot. The mixture is cooled to about -20°C, isolated by filtration from the cold mixture, and dried under reduced pressure to give a dark crystalline solid. The filtrate is discarded. The solids in the extractor

are stirred and the extraction continues with an additional quantity of mixed hexanes to give additional desired product as a dark crystalline solid.

III) Components (A), (C) and (D)

Other materials which are used in preparing the compositions of the Examples are listed in Table 4 below.

Table 4: Components (A), (C) and (D)

Abbreviation	Description
EVA	Ethylene/vinyl acetate copolymer, vinyl acetate content 28 wt %, MI of 6.2 g/10 min; component (C)
PC	Polypropylene copolymer, MI of 25 g/10 min, flex modulus 1000 Mpa determined according to ISO 178 method; component (C)
Zeo4	Zeolite 4A having a pore size of 4Å, average particle size of about 50 microns; component (A)
Zeo13	Zeolite 13X having a pore size of 8Å, average particle size of about 50 microns; component (A)
HD	High Density Polyethylene, homopolymer, MI of 4 g/10 min, density of 0.955 g/ccm; component (C)
PP	Polypropylene, homopolymer, MFI of 3.2 g/10 min, flex modulus of 1450 MPa; component (C)
ATH	Aluminum Trihydrate, average particle size of 5 microns; component (D)
Silica	Synthetic amorphous precipitated silica; specific surface area of 450 m^2/g according to ISO 5794-I, Annex D; average particle size of about 7.5 microns; commercially available as Sipernat® 50 S silica from Degussa Huels, Germany; component (A),
Clay	Calcium montmorillonite, average particle size of 16 microns; commercially available as Nanofill® 918 clay from Suedchemie, Germany; component (A)

III) Compositions

The compositions used in the Examples are listed in Table 5 and prepared as follows:

All components of a particular composition are dryblended. The designations of components (A) – (D) are used as defined hereinbefore, percentages are given as weight percentages based on the total weight of components (A) – (D). The molecular sieve materials of component (A) are used as commercially available and without further treatment. The dryblends are further mixed above melting temperature in a Werner Pfleiderer internal mixer . The blends are formed into films with a thickness of 200 microns on a Collin two roll mill.

Table 5: Compositions

No.	Comp. (A)	Comp. (B)	Comp. (C)	Comp. (D)	DC	DLF
1	Zeo4 1%	ESI-1 99%	none	none	n.a.	n.a.
2	Zeo4 2%	ESI-1 98%	none	none	2.45	0.004
3	Zeo4 4%	ESI-1 96%	none	none	2.57	0.013
4	Zeo4 6%	ESI-1 94%	none	none	n.a.	n.a.
5	Zeo4 10%	ESI-1 90%	none	none	2.89	0.058
6	Zeo4 20%	ESI-1 80%	none	none	3.45	0.185
7	Zeo4 33%	ESI-1 67%	none	none	4.55	0.449
8	Zeo4 50%	ESI-1 50%	none	none	5.94	0.806
9	Zeo13 5%	ESI-1 95%	none	none	2.53	0.043
10	Silica 10%	ESI-1 90%	none	none	n.a.	n.a.
11	Clay 10%	ESI-1 90%	none	none	n.a.	n.a.
12	Zeo4 2%	ESI-2 98%	none	none	2.56	0.014
13	Zeo4 2%	ESI-3 98%	none	none	2.74	0.013
14	Zeo4 2%	ESI-4 98%	none	none	2.37	0.008
15	Zeo4 2%	EPS-1 98%	none	none	n.a.	n.a.
16	Zeo4 10%	ESI-1 50%	EVA 40%	none	2.67	0.087
17	Zeo4 15%	ESI-2 54%	PC 3%	ATH 28%	3.41	0.131
18	Zeo4 6%	none	HD 94%	none	n.a.	n.a.
19	Zeo4 6%	none	PP 94%	none	n.a.	n.a.

Compositions No. 18 and 19 are not within the scope of the present invention.

The DC and DLF data of the compositions are determined according to ASTM D150 test specification using a Hewlett-Packard Impedance/Material Analyzer, Model 4291 B coupled with a Hewlett-Packard HF dielectric material test fixture, Model 16453A. The dielectric properties are measured on film strip samples with a thickness of 200 microns following the instructions in the instrument manual. Frequency scans at 27.12 MHz and ambient temperature are made using a 30 second delay time and point averaging of 64 measurements. A sample of general purpose polystyrene is tested as reference. The DC and DLF for this material are well documented (von Hippel, A. and Wesson, L. G.; *Industrial and Engineering Chemistry*, Vol. 38(11), pp 1121-1129, 1946), the DC should measure in the range between 2.40 and 2.60, and the DLF should measure less than 0.005. If values outside of these ranges are measured the instrument is re-calibrated and/or other corrections are made until acceptable values for the polystyrene reference sample are obtained.

Example 2

Monolayer films (thickness 200 microns) made from the compositions of Example 1 are HF welded by application of dielectric heating. A 1.5 kW BIMA S 361 high frequency generator equipped with a rectangular brass electrode (0.6 cm x 10 cm) opposing a flat brass counter electrode is operated at a frequency of 27.12 MHz. The die is closed with two films of the candidate material in between. Various power settings are used, the electrodes are not preheated. A pressure of 0.7 bar is applied during the welding cycle. After completion of the welding cycle, the die is opened.

The material is subjected to a welding cycle of 8 seconds, with a HF heating phase of 4 seconds and a cooling/holding phase of 4 seconds. For films of a given composition the maximum field strength applied during the heating phase is varied by varying the power setting of the generator in the range of from 20 to 60 scale units. A new sample is made for each power setting.

The resultant welds or seals are examined visually and manually by pulling apart the two films. The welding performance of the films is categorized according to the following criteria – the results are summarized in Table 6.

ns: *no seal*

st: *start seal*; a seal (weld) is formed which is easily peelable without tearing the material forming the seal

s: *seal*; seal has a higher bond strength than '*start seal*' category and is not easily peelable any more. Failure modes are delamination and/or break of material. No weld seam is visible.

GS: *good seal*; the two films cannot be separated along the weld without tearing or breaking either piece of film - no delamination along the weld. Weld seam is clearly visible.

arc: *arcing;* undesirable phenomenon when the power setting is too high; no seal is formed; the films may be partially destroyed.

<u>Table 6:</u> HF Welding Performance at various power settings

Power Setting scale units	Film from Composition No.													
	1	**2**	**3**	**5**	**9**	**10**	**11**	**12**	**14**	**15**	**16**	**17**	**18**	**19**
20				st		ns	ns				st	st		
30	ns	ns	ns	S	ns	st	st	ns	ns	ns	S	S	ns	ns
35											GS	GS		
40	ns	ns	st	S	st	S	S	ns	ns	ns	GS	arc	ns	ns
50	st	st	st	GS	S	GS	arc	st	st	S	arc		arc	arc
52	st			GS	S				S	arc				
54	st/arc	S	S	arc	S	GS		S	S					
56	arc				arc			arc	GS					
58		GS	arc		arc	arc			GS/arc					
60		arc							arc					

Table 7 lists the results obtained with film samples of composition no. 4 for various HF heating times at a constant power setting of 52 scale units and a cooling phase of 4 seconds.

Table 7: HF welding performance at various HF heating times.

Heating Time in seconds	Composition No. 4
1	st
2	st
3	S
4	GS
9	arc

To determine the seal strength at peak, 15 mm strips of film are cut perpendicular to the weld. A Lloyd LR 5K tensile tester is used to peel apart the seal at 180 degrees at a speed of 100 mm/min and grip distance of 35 mm. The seal or bond strength of the welds obtained at the highest possible power setting (without producing an arcing effect) is measured as maximum force at peak and listed in Table 8, as are the failure modes. Pertaining to the failure modes, "D" indicates delamination of the film and "X" breakage at the seal.

Table 8: Seal Strength

Films from Composition No.	Seal Strength Force at Peak (N/15mm)	Failure Mode
1	10	D
2	24	X
3	32	X
5	28	X
9	21	X
10	36	X
11	28	X
12	17	X
13	22	X
14	47	X
15	6	D
16	46	X
17	20	X

Example 3:

A woven polyester fabric (200 g/m^2) is coated on both sides with composition no. 17. On a Collin two roll mill the composition is heated for 5 minutes at set temperatures of 160°C at the rear roll and of 165°C at the front roll. Fabric is fed to the gap at a roll speed of 0.3 m/s. The first side is coated with a 0.8 mm gap, the second side with a 1.5 mm gap. Two of these coated fabrics are HF welded on the equipment described in Example 2 at a power setting of 70 scale units and 4 sec HF heating time. The maximal weld strength is measured as 142 N/15 mm . The sample fails by break of the coating followed by delamination between the coating layer and the polyester fabric.

WHAT IS CLAIMED IS:

1. A HF responsive composition comprising:

(A) from about 2 to about 50 weight percent of at least one molecular sieve material,

(B) from about 30 to about 98 weight percent of at least one interpolymer comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c),

(C) from 0 to about 50 weight percent of another polymer, and

(D) from 0 to about 70 weight percent of an inorganic filler.

2. The composition according to claim 1 wherein component (A) comprises a zeolite.

3 . The composition according to claim 1 or claim 2 wherein the interpolymer (B) is a substantially random interpolymer.

4. The composition according to any of the preceding claims wherein the interpolymer (B) is selected from the group consisting of ethylene/styrene, ethylene/propylene/styrene, ethylene/butene/styrene, ethylene/pentene/styrene, ethylene/hexene/styrene, ethylene/octene/styrene and ethylene/styrene/norbornene.

5. The composition according to any of the preceding claims which has a dielectric loss factor of 0.05 or higher, as measured in a frequency range of from about 13 to 40 MHz and from about ambient temperature to about 80°C.

6. The composition of any of the preceding claims wherein the other polymer (C) is present in an amount of from about 2 to about 50 weight percent and preferably is an ethylene copolymer comprising one or more polar groups.

7. The composition of any of the preceding claims which has undergone a film-forming, a fiber-forming, a profile-forming, a foam-forming, a coating or a molding process.

8. A monolayer or multilayer film comprising a composition according to any of claims 1 to 6.

9. A method for increasing the HF responsiveness of a composition comprising at least one interpolymer comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c),

characterized in that said at least one interpolymer is blended with at least one molecular sieve material.

10. An article of manufacture comprising a composition comprising

(A) from about 2 to about 50 weight percent of at least one molecular sieve material,

(B) from about 30 to about 98 weight percent of at least one interpolymer comprising

(i) polymer units derived from at least one aliphatic olefin monomer having from 2 to 20 carbon atoms; and

(ii) polymer units derived from

(a) at least one vinyl or vinylidene aromatic monomer, or

(b) from at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, or

(c) from a combination of at least one vinyl or vinylidene aromatic monomer and at least one sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomer, and optionally

(d) polymer units derived from at least one ethylenically unsatured polymerizable monomer other than that derived from (a), (b) or (c),

(C) from 0 to about 50 weight percent of another polymer, and

(D) from 0 to about 70 weight percent of an inorganic filler.

11. An article of manufacture according to claim 10 comprising the composition in the form of at least one monolayer or multilayer film.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 02/12021

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08J5/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, CHEM ABS Data, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 99 19398 A (CHEUNG YUNWA W ;DOW CHEMICAL CO (US); GUEST MARTIN J (US)) 22 April 1999 (1999-04-22) cited in the application page 18, line 14-16; claim 1	1,2
A	DATABASE WPI Section Ch, Week 198728 Derwent Publications Ltd., London, GB; Class A25, AN 1987-193933 XP002208677 & JP 62 121759 A (MATSUSHITA ELECTRIC WORKS LTD), 3 June 1987 (1987-06-03) abstract	1-11

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

° Special categories of cited documents :

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
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